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Troposphere-to-stratosphere transport in the lowermost stratosphere from measurements of H_2O, CO_2, N_2O and O_3

E. J. Hintsa,[1] K. A. Boering,[1] E. M. Weinstock,[1] J. G. Anderson,[1] B. L. Gary,[2] L. Pfister,[3] B. C. Daube,[1] S. C. Wofsy,[1] M. Loewenstein,[3] J. R. Podolske,[3] J. J. Margitan,[2] and T. P. Bui[3]

Abstract. The origin of air in the lowermost stratosphere is investigated with measurements from the NASA ER-2 aircraft. Air with high water vapor mixing ratios was observed in the stratosphere at θ~330-380 K near 40 N in May 1995, indicating the influence of intrusions of tropospheric air. Assuming that observed tracer-tracer relationships reflect mixing lines between tropospheric and stratospheric air masses, we calculate mixing ratios of H_2O (12-24 ppmv) and CO_2 for the admixed tropospheric air at θ=352-364 K. Temperatures on the 355 K surface at 20-40 N were low enough to dehydrate air to these values. While most ER-2 CO_2 data in both hemispheres are consistent with tropical or subtropical air entering the lowermost stratosphere, measurements from May 1995 for θ<362 K suggest that entry of air from the midlatitude upper troposphere can occur in conjunction with mixing processes near the tropopause.

Introduction

The exchange of mass between the troposphere and stratosphere is critically important for introducing the source gases involved in ozone destruction (CFCs, N_2O, water, etc.) into the stratosphere and for bringing ozone-rich air down into the troposphere. It is also of interest because the impact of subsonic and future supersonic aircraft exhaust on global ozone levels depends not only on where the emissions are deposited but how long they remain in the stratosphere. More generally, mass transfer across the tropopause is a key process in atmospheric models. At this time, however, large uncertainties remain in the understanding and modeling of cross-tropopause transport.

Since the pioneering work of *Brewer* [1949], it has been thought that air enters the stratosphere in the tropics, where low temperatures at the tropopause dehydrate it to observed stratospheric values, typically 2-6 ppmv. Air then moves up and poleward before descending back into the troposphere in the extratropics (Figure 1). A great deal of subsequent research, both experimental and theoretical [*Holton et al.*, 1995 *and refs. therein*], has largely confirmed this conclusion. However, this simple picture applies only above θ~380 K, the average potential temperature of the tropical tropopause. Outside the tropics, the potential vorticity-based tropopause [*e.g. Holton et al.*, 1995] slopes down more steeply toward the poles than do surfaces of constant potential temperature. The downward slope is particularly steep near the subtropical jet, but continues all the way to high latitudes, where the tropopause may be as low as 300 K. This leads to a distinct region known as the "lowermost stratosphere," located between the local tropopause and the potential temperature of the tropical tropopause, as shown in Figure 1. In contrast, the "overworld" (θ≳380 K, though *Rosenlof et al.* [1997] have recently suggested θ≳450 K, with a "tropically controlled transition region" between 380 and 450 K) is the region above the potential temperature of the tropical tropopause. Air can reach the lowermost stratosphere by three alternative paths (Figure 1): (1) Diabatic descent from the overworld, (2) isentropic (adiabatic) transport from the upper troposphere across the tropopause and into the stratosphere, and/or (3) upward transport across isentropes (diabatic ascent) from the troposphere at midlatitudes.

Previous observations have supported this distinction between the overworld and the lowermost stratosphere. *Dessler et al.* [1995] found H_2O mixing ratios above 10 ppmv in the lowermost stratosphere near 37 N during the Stratospheric Photochemistry, Aerosols and Dynamics Expedition (SPADE). Between the local tropopause and 380 K, measured profiles of H_2O were consistent with a mixture of dry air from the overworld and moister air that had been transported isentropically from the tropical upper troposphere. Aircraft transects of the subtropical tropopause [*Folkins and Appenzeller,* 1996], *in situ* measurements in the lowermost stratosphere [*Kritz et al.*, 1991], and model calculations [*Yang and Pierrehumbert,* 1994; *Chen,* 1995] have also

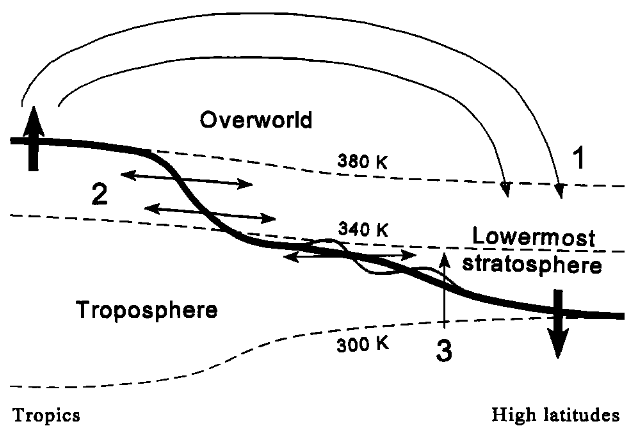

Figure 1. Schematic of the atmosphere, with the thick line denoting the average tropopause. The lowermost stratosphere lies between the tropopause and ~380 K; above that is the overworld. Thick arrows show stratosphere/troposphere exchange through the Brewer-Dobson circulation, and the labeled arrows show pathways by which air may enter the lowermost stratosphere. Path 1 represents descent from the overworld, 2 represents transport along isentropes from the upper troposphere, and 3 represents diabatic ascent at midlatitudes. Note that path 2 can occur at midlatitudes as well as the subtropics, due to the slope of the tropopause (shown schematically by the thin curved line about the average tropopause) with respect to isentropes.

[1]Harvard University
[2]Jet Propulsion Laboratory
[3]NASA Ames Research Center

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suggested that path 2 is active. In contrast, *Poulida et al.* [1996] observed a thunderstorm in North Dakota (47 N) with convective outflow into the stratosphere. They calculated that significant amounts of water were transported locally into the stratosphere through path 3. In recent papers, *Bregman et al.* [1997] and *Lelieveld et al.* [1997] observed tropospheric air in the lowermost stratosphere, based on aircraft measurements of CO and other species. The air was thought to have mixed into the stratosphere at mid-latitudes, by convective or frontal activity.

In this study, simultaneous observations of H_2O, CO_2, N_2O, and O_3 in the lowermost stratosphere are used to discriminate between the various pathways into the stratosphere. Air passing through the tropical tropopause is very dry, whereas air entering by other paths will be wetter, reflecting the higher minimum temperatures encountered. Water vapor is a very sensitive tracer for this type of exchange, as it can be an order of magnitude or more higher in the upper troposphere than in the stratosphere. Thus, it can distinguish path 1 from paths 2 and 3 (Figure 1). In contrast, observations of CO_2 allow us to distinguish paths 1 and 2 originating in the tropics/subtropics from path 3 (originating at midlatitudes). Since CO_2 mixing ratios are in general different in the tropical/subtropical upper troposphere than in the midlatitude upper troposphere [*e.g., Nakazawa et al.*, 1991] because of spatial and temporal variations in its sources and sinks, CO_2 can be used as a probe of the origin of air that is transported from the troposphere to the stratosphere. Ozone and N_2O serve as long-lived tracers with respect to cross-tropopause exchange and provide a framework with which to evaluate the water and CO_2 measurements.

Measurements

During the Stratospheric TRacers of Atmospheric Transport (STRAT) campaign to study transport in the lower stratosphere, the NASA ER-2 aircraft flew from Ames Research Center (37 N, 122 W) in May 1995 with a small subset of instruments measuring long-lived tracers. Two of the flights (950515 and 950517, in YYMMDD format) occurred in a "stairstep" configuration, in which the aircraft flew approximately half-hour horizontal flight tracks at different altitudes, with the lowest tracks very close to the tropopause. Water vapor was measured by Lyman-α photofragment fluorescence [*Weinstock et al.*, 1994], CO_2 by non-dispersive infrared absorption [*Boering et al.*, 1994], N_2O by tunable diode laser absorption [*Podolske and Loewenstein*, 1993], and O_3 by UV absorption [*Proffitt and McLaughlin*, 1983]. Tropopause locations were determined from remote-sensing Microwave Temperature Profiler (MTP) [*Denning et al.*, 1989] and *in situ* Meteorological Measurement System (MMS) data [*Scott et al.*, 1990] using the WMO definition (the lowest altitude for which dT/dz > −2 K/km for 2 km), with typical uncertainties of 0.1-0.3 km.

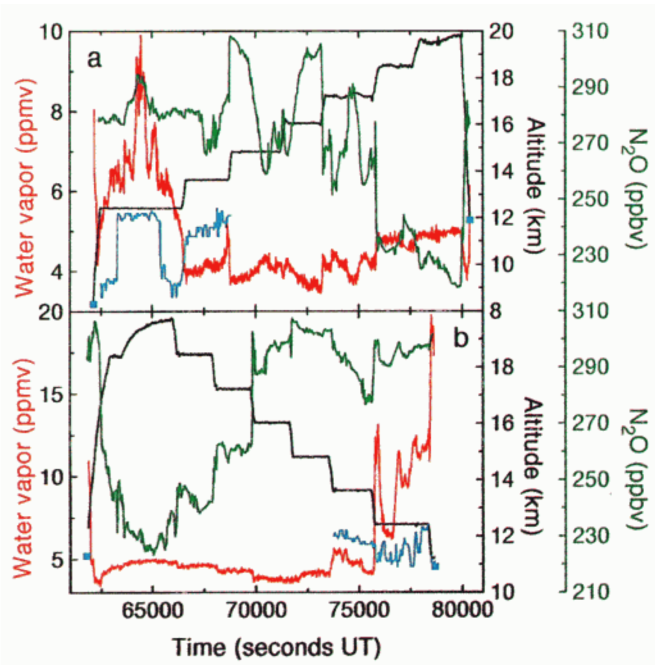

Figure 2. Time series of ER-2 altitude (black), tropopause altitude (blue), H_2O (red), and N_2O (green) in the stratosphere for (a) 950515 and (b) 950517. Altitudes below ~14 km (~383 K) are in the lowermost stratosphere; blue squares show where the ER-2 crossed the tropopause. The latitude range on each flight track is approximately 37-41 N.

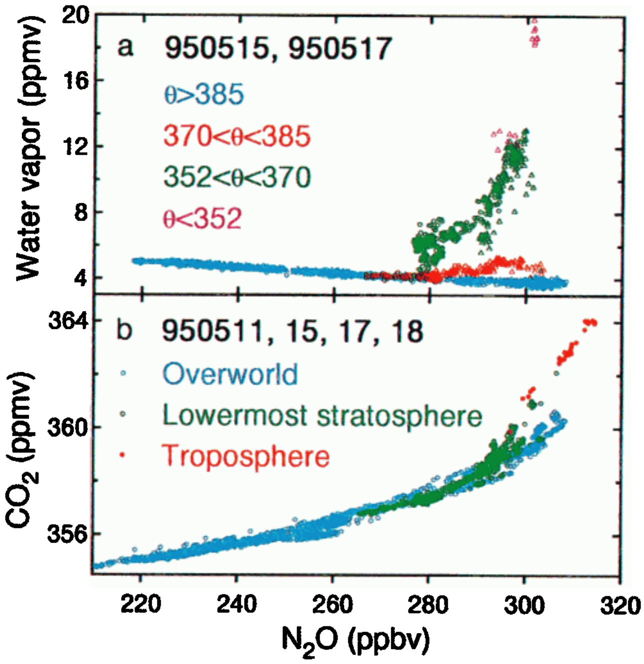

Figure 3. (a) Correlation plot of stratospheric H_2O vs. N_2O for 950515 (○) and 950517 (△). The large deviations (upper right) from the typical tight correlation were measured on the lowest flight tracks (352-370 K, ~12.4 km, green) with smaller deviations obvious up to 385 K (~14 km, red). For 950517, data between the tropopause and 352 K are shown in magenta; for 950515, no N_2O data are available below 352 K. (b) CO_2-N_2O correlation plot, showing the smooth transition from the lowermost stratosphere (green) to the troposphere (red). Data in the overworld are shown in cyan throughout.

Water vapor and N_2O on the stratospheric parts of the stairstep flights are shown in Figure 2. Regions of high H_2O (>8 ppmv) are clearly evident on the lowest flight tracks, associated with N_2O lower than the tropospheric value of ~315 ppbv. H_2O and N_2O are tightly correlated above 385 K (Figure 3a), similar to SPADE data [*Hintsa et al.*, 1994] (most of which were obtained in the overworld), but show deviations toward higher water at lower altitudes. High levels of ozone (300-550 ppbv) confirm the stratospheric character of this air with elevated H_2O.

The flight of 950515 began with the tropopause at 8.3 km (314 K) based on MMS data, but as the ER-2 flew north at 12.4 km the tropopause rose until it was within 364±12 m of the aircraft at the northernmost point, where H_2O peaked at almost 10 ppmv (Figure 2a). The ER-2 then retraced its path south at the same altitude, leading to a symmetric profile. However, as determined by MTP and consistent with other tracer measure-

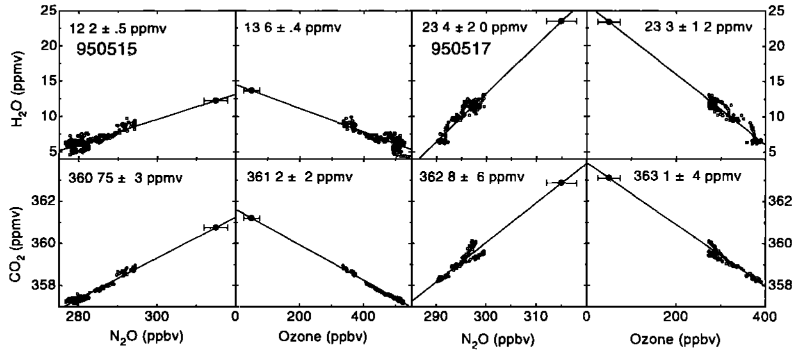

Figure 4. Correlation plots for the lowest flight tracks (950515, left; 950517, right) and linear fits (lines). Extrapolated tropospheric H_2O and CO_2 are shown as solid circles and listed in each figure, with error bars from tropospheric boundary conditions.

ments, the aircraft was always in the stratosphere. Above θ=370 K (12.8 km), water vapor was only slightly higher than typical, and the H_2O-N_2O correlation was very tight except for some of the data at 380-383 K (~13.7 km, shown in red in Figure 3a).

On 950517, H_2O up to 385 K (14.4 km) was higher than would be predicted from the overworld H_2O-N_2O correlation. The tropopause was located at 10.9 km (332 K) on descent, where H_2O increased to almost 20 ppmv before the ER-2 reentered the troposphere (Figure 2b).

Analysis and Discussion

The observation of 9-20 ppmv of H_2O in the stratosphere clearly indicates the entry of air other than via path 1 (Figure 1). The moist air masses were within 2.5 km of the tropopause (with the wettest air even closer), low enough in altitude to have entered the stratosphere in the midlatitudes or subtropics. For the lowest horizontal flight tracks (Figure 3a, green), the H_2O-N_2O relationship is roughly linear, suggesting a mixing line between two air masses on an isentropic surface in the stratosphere, one having descended from the overworld, the other from the upper troposphere. From this premise, we calculate the composition of both air masses (the "end members" of the mixing line) and the fractional amounts of each, determine the likely origin of the tropospheric air from CO_2 data, and try to elucidate the mechanism by which it entered the stratosphere.

We use elevated H_2O to isolate regions in the stratosphere influenced by air that did not cross the tropical tropopause. We then extrapolate the mixing lines in the H_2O-N_2O, H_2O-O_3, CO_2-N_2O, and CO_2-O_3 correlations to tropospheric boundary conditions for N_2O and O_3 in order to determine the entry level mixing ratios of H_2O and CO_2 (Figure 4). The tropospheric boundary conditions (including atmospheric variability and measurement uncertainties) are N_2O=315±3 ppbv, O_3=50±25 ppbv and high but variable H_2O. Air in the overworld has low H_2O (2-6 ppmv) but variable (and lower) N_2O and variable (and higher) O_3. CO_2 varies systematically in both regions, with differences between the midlatitude and the tropical/subtropical upper troposphere as noted earlier. As a check, O_3-N_2O correlations in the regions with elevated H_2O were examined and found to extrapolate correctly to their boundary conditions.

For 950515, an extrapolation of the H_2O-N_2O relationship at θ=352-364 K (12.4 km; ~0.5 km above the local tropopause) to N_2O=315 ppbv results in an H_2O entry level of 12.2 ppmv for the tropospheric end member. Regressing H_2O vs. O_3 gives an entry level of 13.6 ppmv. Similarly, CO_2 regressed against N_2O and O_3 for the same range of θ both give a CO_2 entry level of ~361 ppmv. This value suggests a tropical or subtropical origin for the tropospheric air, since subtropical CO_2 was in this range at that time, as was CO_2 in air entering the overworld in the tropics [*Boering et al.*, 1996], whereas CO_2 measured in the upper troposphere at Ames was at least 363-364 ppmv.

For 950517, the H_2O-N_2O correlation at θ=352-356 K (12.4 km; 0.5-1.5 km above the local tropopause) leads to an H_2O entry level of 23.4 ppmv (23.3 ppmv from the H_2O-O_3 correlation), much higher than for 950515. The CO_2 correlations diverge slightly toward higher N_2O and lower O_3, but the extrapolated values of tropospheric CO_2 are 362.8 ppmv (from N_2O) and 363.1 ppmv (from O_3), very close to local tropospheric values and too high for the tropics/subtropics, suggesting the entry of midlatitude air into the lowermost stratosphere. Additionally, CO_2-N_2O correlations at θ<354 K during this deployment (Figure 3b) show a continuous transition from stratospheric to local tropospheric air, with low N_2O values (290-305 ppbv) sampled in the troposphere, consistent with two-way exchange across the midlatitude tropopause.

The observed correlations from 950515 are consistent with tropospheric air being transported from the tropics or subtropics across the steeply sloped subtropical/midlatitude tropopause into the lowermost stratosphere (path 2 in Figure 1), while data from 950517 are consistent with exchange between the midlatitude upper troposphere and the lowermost stratosphere (path 3, or path 2 at midlatitudes, see below). For both flights, the mixed air masses at 352-364 K (12.4 km) in the lowermost stratosphere range from about 5 to 55% tropospheric air, with the balance composed of air characteristic of the overworld.

From these and other ER-2 measurements, including data from SPADE [*Dessler et al.*, 1995], the southern hemisphere [*Tuck et al.*, 1997], and subsequent STRAT deployments, it appears that this phenomenon of tropospheric air mixing directly into the lowermost stratosphere is ubiquitous. The majority of CO_2 data suggests that this air is predominantly from the tropics/subtropics, though for 950515 the actual cross-tropopause transport may have occurred at midlatitudes. Weather patterns just prior to 950515 transported subtropical tropospheric air on the 355 K surface in the Pacific to 50-60 N. Isentropic back trajectories ending along the 950515 ER-2 flight track in the lowermost stratosphere showed air parcels traveling over this subtropical air mass near 50 N. Thus, the observations are consistent with tropical/subtropical air moving north and entering the stratosphere at midlatitudes, where back trajectories passed very close to the tropopause. For 950517, both the extrapolated CO_2 values and back trajectories indicate that midlatitude tropospheric air had mixed into the lowermost stratosphere.

Cross-tropopause transport at midlatitudes can occur either along or across isentropes, since the local tropopause is in general not parallel to isentropes and can be extremely sloped. For example, on 950515 the tropopause was at 8.2 km (314 K) near 37.5 N and at 12.1 km (~350 K) near 40.5 N. Small-scale wave activity or two-dimensional (horizontal) turbulence could then have mixed air quasi-isentropically between the troposphere and the stratosphere. No strong convection was observed along 10-day back trajectories for the flights of 950515 and 950517. Therefore, the observed mixing is likely to have been along isentropes, or nearly so, with no contribution from convection. This is simply path 2 (Figure 1) for $\theta \leq 360$ K occurring at mid and high latitudes on isentropes which cross the tropopause.

The saturation temperatures consistent with the extrapolated water vapor entry levels provide further clues as to the location of transport into the stratosphere. For 950515, the extrapolated H_2O mixing ratio was 13 ppmv, corresponding to a saturation temperature of 200-201 K on the 354 K isentrope. For 950517, the extrapolated mixing ratio of 23.4 ppmv corresponds to 205 K. In National Centers for Environmental Prediction data, temperatures of 205-206 K were present at midlatitudes on May 13-17 over the eastern Pacific on the 354 K surface, with 200 K reached on May 5-7. Minimum temperatures on the 354 K surface were all in the subtropics and midlatitudes (20-40 N), close to the tropopause. Thus, air could have been dehydrated in the extratropics to the extrapolated values.

The fate of tropospheric air mixed into the lowermost stratosphere is almost certainly diabatic descent back to the troposphere. Anomalously high water was observed only at $\theta \leq 385$ K, and is unlikely to be transported to higher altitudes given the vertical stability of the stratosphere. Furthermore, the observed deviation of H_2O from its expected correlation with N_2O (Figure 3a, cyan) generally decreased with altitude above the tropopause. Nonetheless, a small amount of upward mixing could have a slight impact (~0.1 ppmv) on the stratospheric hydrogen budget. *Rosenlof and Holton* [1993] have calculated a turnover time of 5-8 months for transport from the lowermost stratosphere into the troposphere. Accordingly, air in the lowermost stratosphere, and therefore aircraft effluent, will be predominantly transported downward. However, as suggested by *Chen* [1995], some fraction of air in the lowermost stratosphere could also be transported laterally into the tropical upper troposphere, where it may affect chemistry and the radiative balance.

This paper illustrates a tracer-tracer method for identifying (1) regions of the lowermost stratosphere influenced by transport across the extratropical tropopause and (2) the tropospheric "end-member" air as being either tropical/subtropical or midlatitude in origin, both important steps toward quantifying stratosphere/troposphere exchange. Further work is continuing to determine the seasonality of transport into the lowermost stratosphere, and mixing between this region and the tropics.

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J. G. Anderson, E. J. Hintsa, and E. M. Weinstock, Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA 02138. (e-mail: hintsa@huarp.harvard.edu)

K. A. Boering, B. C. Daube, and S. C. Wofsy, Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA 02138.

B. L. Gary and J. J. Margitan, Jet Propulsion Laboratory, Pasadena, CA 91109.

T. P. Bui, M. Loewenstein, L. Pfister, and J. R. Podolske, NASA Ames Research Center, Moffett Field, CA 94035.

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